COAXIAL PULSED CORONA REACTOR FOR TREATMENT OF HAZARDOUS GASES

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Abstract

The destruction of hazardous gaseous chemicals has in the past been effectively accomplished using thermal techniques. Pulsed corona reactors are promising candidates for more efficient destruction of hazardous gases using a non-thermal electrical discharge at atmospheric pressure. The energy required for chemical destruction is deposited in the medium by highly energetic electrons present near the streamer head while direct heating of the neutral gas molecules is avoided. The current work emphasizes the construction of an electrically efficient, low cost pulsed corona reactor capable of treating a flow rate of 4 liters/minute. Determining optimum pulse parameters for the discharge and scaling the device to higher flow rates will be the primary focus of future work, with specific emphasis being placed on the use of fast-rising pulses (nanoseconds) of limited pulse-width (10's to 100's of nanoseconds). A testbed for chemical analysis has also been assembled and preliminary results include the electrical efficiency of the device and the chemical destruction efficiency when challenged by small concentrations of hazardous gases in ambient air.

Introduction

Recent attention to (and public awareness of) environmental pollution involving gaseous wastes has necessitated the development of an effective means of remediation. The use of non-thermal plasma reactors for the destruction of these hazardous gaseous chemicals shows promise of being highly efficient when compared to thermal methods [1,2,3]. The success of a non-thermal plasma approach is contingent upon the creation of a discharge at atmospheric pressure in which the majority of the energy is spent on producing energetic electrons rather than on heating of neutral gas molecules. The radicals produced by collision of these energetic electrons with the background gas have a relatively long lifetime and can react selectively with the contaminant molecules leading to their eventual neutralization as a threat. One non-thermal plasma device of particular interest to the Navy is the Pulsed Corona Reactor (PCR). In this device, short, high-voltage pulses are rapidly applied to a coaxial electrode arrangement in order to produce multiple streamer discharges in a flowing ambient gas volume. Previous work [4,5] has concentrated on small-scale laboratory demonstrations using rotating spark gaps and resistive elements to control charging of the primary energy store as well as the pulse shape. In addition, only minute flow rates were considered. Despite these limitations, significant chemical destruction efficiencies of small concentrations of pollutants such as toluene (C_7H_8), dichloromethane (CH_2Cl_2), and dichlorodifloromethane (CF_2Cl_2-Freon 12) were demonstrated at moderate voltages, flow rates of 1 liter/minute (lpm), and repetition-rates which were limited to less than 300 Hz. Key issues in the development of a viable system, however, will necessarily involve *both* the electrical and chemical efficiency of the device.

The current effort was initiated to address first the question of electrical efficiency followed by maximization of the chemical destruction efficiency through manipulation of pulse parameters, namely the risetime, peak voltage and pulse width. It is our intention to maximize the voltage (or E/N) which can be applied to the reactor through the application of nanosecond risetime pulses of limited pulsewidth (10-100 ns). Since only short pulses of electrons are needed to create long-lived radicals, longer pulses are deemed unnecessary. The reactor flow-rate will be increased through high rep-rate hydrogen spark-gap switching. Additional attention will be given to minimizing the physical size and cost of the device through a compact coaxial design.

Experimental Apparatus

A schematic diagram of the pulsed corona reactor is given in Figure 1. A Maxwell CCDS 8 kJ/s constant-current switching power supply is used to quickly charge the primary capacitive store, C_1. The supply is remotely controlled via high voltage enable, high voltage inhibit and voltage level set signal lines. The capacitance C_1 can consist of either a pulse-forming line or a discrete capacitance, depending on the desired pulse parameters. The values of capacitance shown were necessary to obtain sufficient regulation from the supply. The power rating of the supply is not well suited for such a small reactor, but is needed for eventual scaling of the device. A hydrogen spark gap operating in a self-break mode is used to initiate the fast-rising pulse which is applied to the reactor. By using hydrogen gas as the switching media in the peaking gap, operation of up to 1 kHz in the self-break mode will be possible [6]. The switch housing is made out of Lexan and the electrodes are made out of brass. Gap spacing and switch pressure can be adjusted to control the breakdown voltage. The switch is contained within a 3" diameter copper tube and connects coaxially with the reactor chamber. The voltage is resonantly applied to the reactor upon closure of the peaking switch.

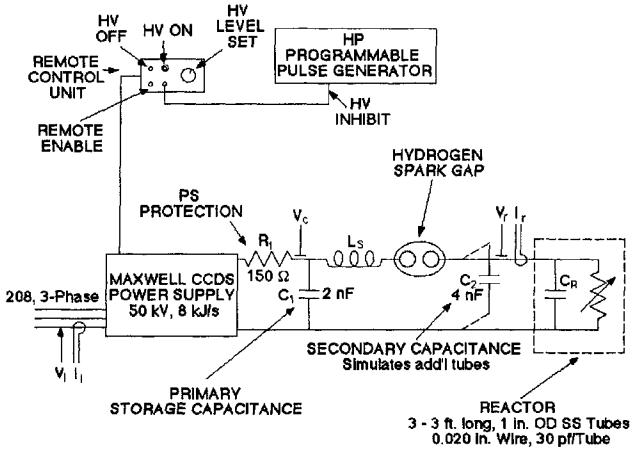

Figure 1. The schematic diagram for the pulse corona system. The circuit elements from the power supply to the reactor are coaxial, V_r and V_c are capacitive divider voltage probes, and I_r and I_i are Pearson current monitors.

Figure 2 shows a cross-sectional view of an individual reactor module. The module consists of a cylindrical stainless steel tube with a 0.9" inside diameter. A 0.020" diameter stainless steel wire is positioned in the center of the tube by a tensioning screw and special crimp located at either end of the chamber. The feedthroughs are fabricated out of Teflon. All seals are made with

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14. ABSTRACT
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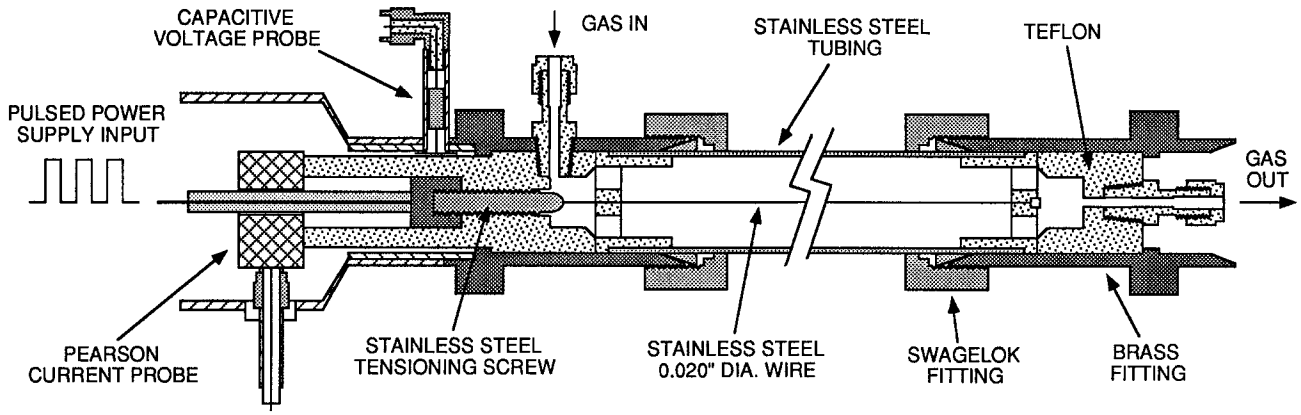

Figure 2. The plasma reactor section of the pulse corona system. The gas flow is in contact with stainless steel and teflon only. The placement of the fast diagnostics is also indicated.

Teflon and Swagelok fittings so that the hazardous gas only comes in contact with the stainless steel and teflon. Removal of the brass fitting on the downstream side of the tube allows visible access to the reactor interior during operation. At present, the reactor consists of three such tubes, each 3-ft. in length, connected in parallel. Each reactor has a capacitance of 30 pF. The entire geometry of the pulsed power supply and reaction chamber is coaxial so that problems with EMI and personnel safety are avoided.

A measurement of the voltage applied to the reactor is obtained with a fast capacitive divider probe located at the reactor input. The total reactor current is monitored with a Pearson probe capable of a few-nanosecond risetime operation. All signals are recorded simultaneously on a HP54111 digitizing oscilloscope with the acquired data being transferred to a computer for subsequent analysis.

The destruction efficiency of the pulsed corona reactor for a particular gaseous chemical was measured using the apparatus of Figure 3. A predetermined quantity of the hazardous compound is injected into a 85 liter gas bag. The bag is then filled at a known rate and for a given time period in order to yield a particular concentration of hazardous compound in parts per million (ppm). A three-way valve connects the bag to a mass flow meter (MFM) and the PCR, with a plug valve being used to control the flow rate through the system. A measurement of the relative pollutant concentration at the outlet of the reactor is made with a micro gas chromatograph (μGC). Software support for the μGC allows the data to be stored and analyzed on a dedicated computer.

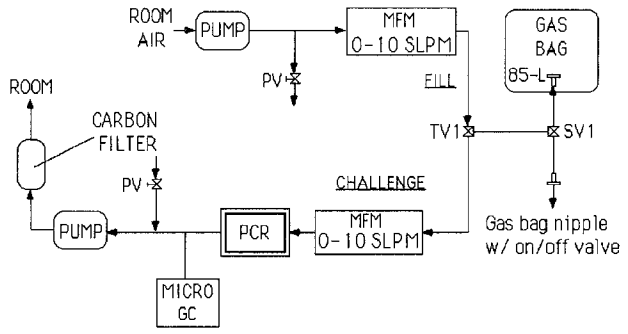

Figure 3. The challenge apparatus used to obtain the percent destruction of a small concentration of target gas (100's of ppm) in air. A gas bag is used to obtain a precise concentration and a micro gas chromatograph records the percent destruction.

Experimental Results

The electrical characteristics of the reactor were measured as shown in the correlated waveforms of Figure 4. Figure 4a shows the resonant voltage transfer to the reactor and secondary capacitance from the prime store. These measurements were obtained with a flow rate of 4 lpm in ambient air and a rep-rate of 200 Hz. The risetime of the pulse is about 25 ns. The voltage decays to the corona onset threshold of approximately 10 kV after the discharge event. The voltage difference between the beginning and end of the voltage waveform is attributable to a secondary discharge event. This subsequent discharge event is believed to be a DC corona which leads to an additional decay in the voltage to a secondary threshold of about 8.5 kV. Verification of this explanation is provided by using a smaller diameter wire (0.010") which resulted in a lower secondary threshold. As this discharge occurs on a much longer timescale (>10 μs), it is considered undesirable. In fact, a noticeable increase in gas temperature results when operating with the 0.010" diameter wire. Figure 4b shows the total current flowing into the three reactor tubes. As can be seen, the pulse is made up of two components: the displacement current between the two capacitors C_1 and C_R, and the actual streamer current of the reactor. The pulse duration is less than 200 ns, and the pulse amplitude is 135 A for an applied voltage of 14 kV. The resistance of the discharge is given in Figure 4c. The resistance of the multichannel streamer discharge falls to approximately 75 Ω and then exponentially increases. Figure 4d shows the power into the reactor where the energy deposited in the discharge is approximately 110 mJ.

At a repetition-rate of 200 Hz, the above waveforms were statistically averaged to obtain the power input to the reactor. Similarly, the average power required from the three-phase line input to the power supply was measured. A comparison of the two measurements indicates an electrical efficiency of greater than 90% for the device.

The chemical destruction efficiency was measured by challenging only one of the three parallel tubes of the reactor. A flow rate of 4 lpm was maintained in each tube corresponding to a residence time of 1.5 seconds. At a reactor voltage of 14 kV and a repetition-rate of 100-200 Hz, the destruction efficiencies for three hazardous compounds are shown in Figure 5. Initially, the flow from the gas bag out to the PCR and μGC is allowed to stabilize, after which the PCR is enabled at the repetition-rates indicated. All pollutant concentrations were near 250 ppm. A maximum destruction efficiency for toluene, dichloromethane, and dichlorodifluoromethane was 32%, 10%, and 4% respectively. Freon-12 is the most difficult of the three to destroy.

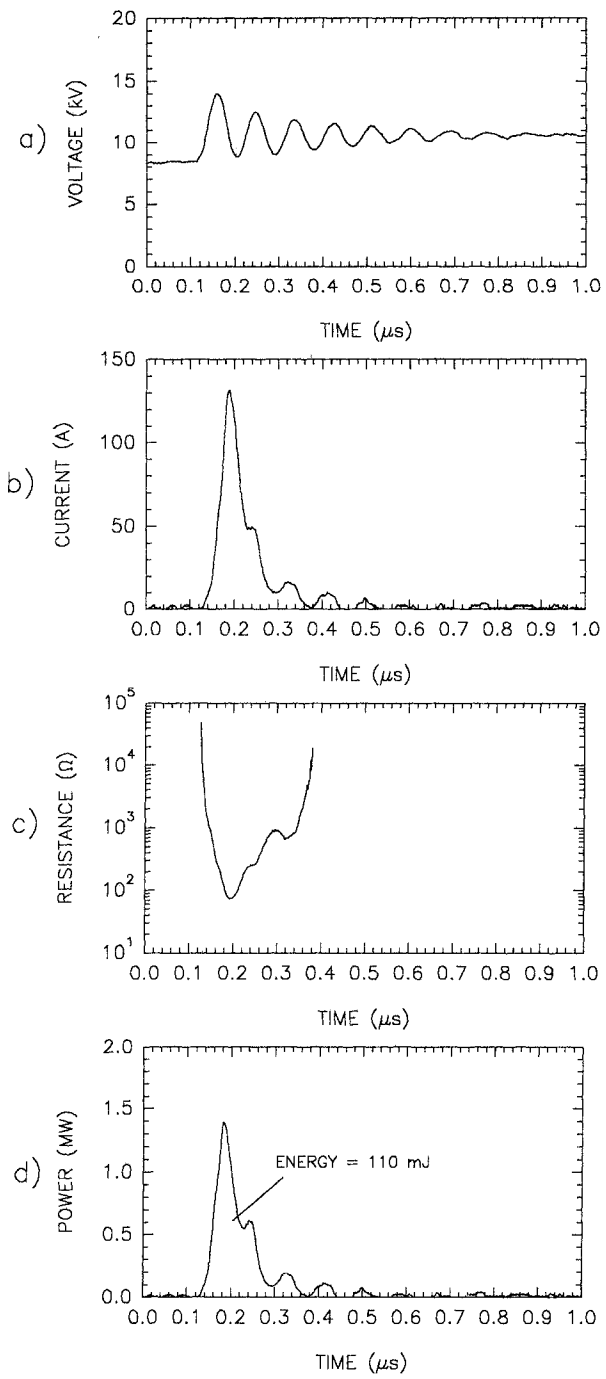

Figure 4. The electrical characteristics of the pulse corona reactor where: a) is the voltage applied to the reactor when the spark gap fires as measured by a fast capacitive divider probe, b) is the current in the corona discharge measured with a Pearson current monitor, c) is the total resistance of the corona streamers, and d) is the power delivered to the discharge (the energy is the area under the power vs. time curve).

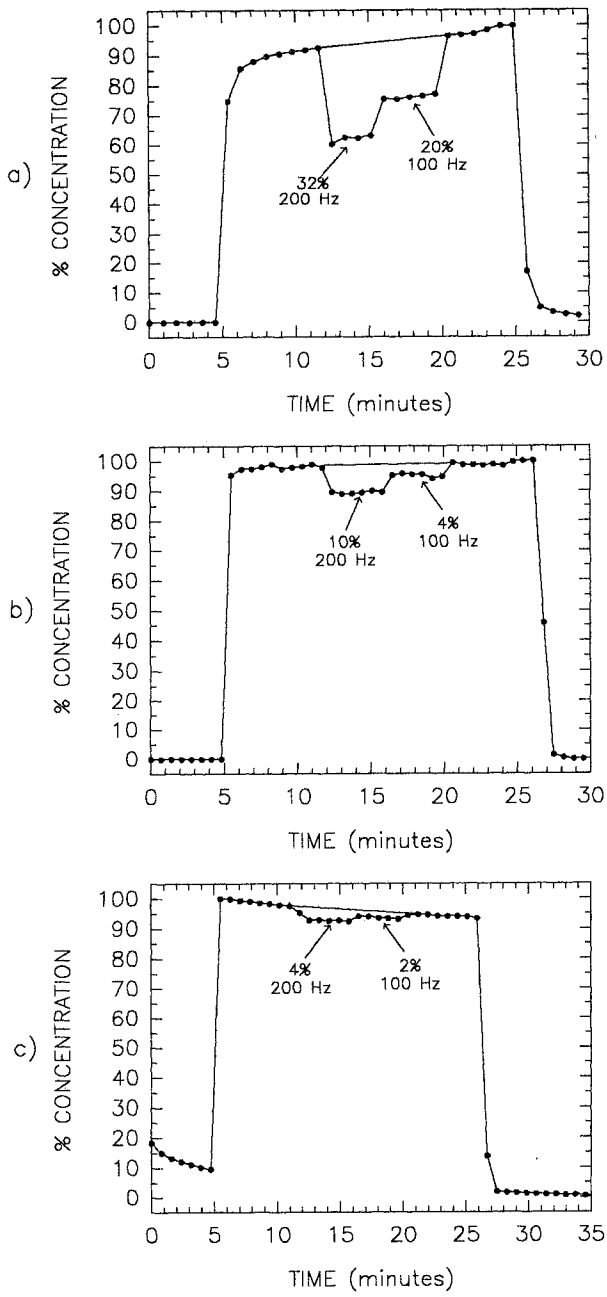

Figure 5. The percent concentration versus time obtained from the micro-gas-chromatograph measured at the output of the reactor where: a) is for 270 pmm toluene (C_7H_8), b) is for 230 ppm dichloromethane (CH_2Cl_2) and c) is for 240 ppm dichlorodifluoromethane (CF_2Cl_2).

Discussion

The above results serve as a baseline for further improvements in the performance of the device. Improved reproducibility of the amplitude of the voltage pulse applied to the reactor will result in a greater operating range for the device. Improved regulation from the power supply will be a natural consequence of scaling the reactor to a larger size (i.e. requiring larger C_1). Operating the hydrogen spark gap at higher pressures (100 psi) will help reduce the statistical jitter in the self-break voltage of the peaking gap, although triggering of the gap can be implemented if necessary. This jitter has been linked to the inability of the power supply to inhibit its high voltage output upon breakdown of the peaking gap,

thereby leading to spurious arcing in the reaction chamber at higher voltages. A reduction in the risetime, as well as a reduction in the pulsewidth, will be realized by replacing C_1 with a pulse forming line. C_2 will be replaced with the capacitance of multiple reactor tubes so that flow rates of up to 500 lpm can be processed. The reactor geometry is capable of supporting approximately 35 kV before arcing occurs so that marked improvement in the chemical destruction efficiency is expected. In addition, taking full advantage of the high rep-rate capability of the power supply and hydrogen switch will no doubt lead to increased chemical destruction efficiency and reactor flow rates.

Conclusions

An electrically efficient pulsed power supply has been constructed using a constant current switching supply and single hydrogen gas switch. Stable operation has been demonstrated at voltages up to 14 kV at 200 Hz and the chemical destruction efficiencies for toluene, methylene chloride, and Freon-12 have been measured. From this baseline, maximization of the device's chemical destruction efficiency is being pursued through maximization of the operating voltage and repetition-rate, and minimization of the risetime and pulse width.

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